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D E C I S I O N
of 16 January 2002

Case Number: T 0677/98 - 3.3.1

Application Number: 90904062.8

Publication Number: 0455748

IPC: C07C 19/08

Language of the proceedings: EN

Title of invention:

Manufacture of 1,1,1,2-tetrafluoroethane

Patentee:

E.I. DU PONT DE NEMOURS AND COMPANY

Opponent:

AUSIMONT S.p.A.

Headword:

1,1,1,2-tetrafluoroethane/E.I. DU PONT DE NEMOURS

Relevant legal provisions:

EPC Art. 54(1)(2), 56

Keyword:

"Main request - first and second auxiliary request - inventive step (no) - obvious alternative solution"

Decisions cited:

-

Catchword:

-



Case Number: T 0677/98 - 3.3.1

D E C I S I O N
of the Technical Board of Appeal 3.3.1
of 16 January 2002

Appellant: AUSIMONT S.p.A.
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Decision under appeal: Interlocutory decision of the Opposition Division
of the European Patent Office posted 27 April
1998 concerning maintenance of European patent
No. 0 455 748 in amended form.

Composition of the Board:

Chairman: P. P. Bracke
Members: P. F. Ranguius
S. C. Perryman

Summary of Facts and Submissions

I. The Appellant (Proprietor of the patent) lodged an appeal against the interlocutory decision of the Opposition Division which maintained the European patent No. 0 455 748 (European patent application No. 90 904 062.8) in the form as amended (sixth auxiliary request filed before the Opposition Division) pursuant to Article 102(3) EPC.

II. The patent was granted with thirteen claims, independent Claim 1 (the sole independent claim) reading:

"1. A process for the manufacture of 1,1,1,2-tetrafluoroethane by the reaction of HF and trichloroethylene in the presence of 2-chloro-1,1,1-trifluoroethane and a catalyst at elevated temperature to form a mixture comprising 1,1,1,2-tetrafluoroethane, 2-chloro-1,1,1-trifluoroethane and optionally other organic by-products, wherein

- the 2-chloro-1,1,1-trifluoroethane in said mixture is recycled from the mixture to the reaction zone along with additional trichloroethylene in a molar amount at least equal to the molar amount of 1,1,1,2-tetrafluoroethane recovered from the mixture and with additional HF in a molar amount from 3 to 30 times the molar amount of trichloroethylene, and
- said reaction is conducted in a single reaction zone at a temperature of 300 to 500°C and at a contact time of 0.1 to 60 seconds and in the presence of a catalyst selected to form a mixture comprising 1,1,1,2-tetrafluoroethane and 2-chloro-1,1,1-trifluoroethane and less than 10 percent by

weight of said other organic by-products; said catalyst being a catalyst composition comprising at least one of trivalent chromium, a Group VIII, Group VIIB, Group IIIB or Group IB metal and metals having an atomic number from 58 to 71".

III. The opposition sought revocation of the patent in suit in its entirety, in particular on the grounds that the subject matter of Claim 1 lacked novelty or did not involve an inventive step. The following documents were *inter alia* cited.

(1) US-A- 4 792 643

(6) US-A- 4 158 675

(8) US-A- 4 129 603

IV. The Opposition Division held that, starting from document (1) as the closest state of the art, the problem underlying the patent in suit might be seen in manufacturing 1,1,1,2-tetrafluoroethane (HFC-134a) by the reaction of HF and trichloroethylene in the presence of a catalyst without the need for two reactors and where less than 10 wt% of organic by-products are produced. The claimed solution was obvious in view of the disclosure of document (1) for the following reasons:

- Examples Nos. 13 and 14 of document (1) disclosed the manufacture of HFC-134a with trivalent chromium catalyst in reaction conditions similar to those of the claimed invention. Furthermore, the general disclosure of this document included the possibility of recycling 2-chloro-1,1,1-

trifluoroethane (HCFC-133a), the last intermediate before HFC-134a, for further reaction. Carrying out the reaction in a single reactor was, therefore, considered as obvious.

- Furthermore, adding trichloroethylene and HF in amounts as defined in Claim 1 was considered as self-evident for maintenance of the reaction.

V. At the oral proceedings before the Board which took place on 16 January 2002, the Appellant filed

- as main request, a set of thirteen claims, Claims 1 to 12 being the claims as granted and Claim 13 being amended to read:

"13. The process of claim 1 wherein other organic by-products in said mixture are recycled".

- as first auxiliary request, the set of Claims 1 to 12 as granted, Claim 13 of the main request being deleted.

- as second auxiliary request, a set of eleven claims, Claim 1 reading:

"1. A process for the manufacture of 1,1,1,2-tetrafluoroethane by the reaction of HF and trichloroethylene in the presence of 2-chloro-1,1,1-trifluoroethane and a catalyst at elevated temperature to form a mixture comprising 1,1,1,2-tetrafluoroethane, 2-chloro-1,1,1-trifluoroethane and optionally other organic by-products, wherein

- the 2-chloro-1,1,1-trifluoroethane in said mixture is recycled from the mixture to the reaction zone along with additional trichloroethylene in a molar

amount at least equal to the molar amount of 1,1,1,2-tetrafluoroethane recovered from the mixture and with additional HF in a molar amount from 3 to 30 times the molar amount of trichloroethylene, and

- said reaction is conducted in a single reaction zone at a temperature of 300 to 500°C and at a contact time of 0.1 to 60 seconds and in the presence of a catalyst selected to form a mixture comprising 1,1,1,2-tetrafluoroethane and 2-chloro-1,1,1-trifluoroethane and less than 10 percent by weight of said other organic by-products; said catalyst being a catalyst composition comprising trivalent chromium metal".

VI. In a communication accompanying the summons to oral proceedings, the Board introduced a new document as common general knowledge:

(16) Hawley's Condensed Dictionary, 11th edition, 1987, page 998,

regarding the definition of the term "recycling".

VII. The Appellant's submissions both in the written proceedings and at the oral proceedings before the Board can be summarised as follows:

- Document (1) related to a process for the conversion of trichloroethylene to 1,1,1,2-tetrafluoroethane (HFC-134a) involving a catalyst prepared by co-depositing a hexavalent chromium oxide and a transition metal on alumina. The subject matter of the claimed invention was new over document (1) given that it did not disclose

the same catalyst system and the process was not a continuous process with recycling the 1,1,1-trifluoro-2-chloroethane (HCFC-133a) to a single zone with specific amounts of trichloroethylene and HF.

- In view of document (1) as the closest state of the art, the technical problem to be solved might be seen in the provision of a process for economically manufacturing HFC-134a in a high amount and with a minor amount of organic by-products. Contrary to the Opposition Division's view, document (1) did not teach the suitability of any of the catalysts comprising trivalent chromium. Indeed, said document taught, in its examples Nos. 13 and 14, that trivalent chromium was inferior to the catalysts based on hexavalent chromium oxide and titanium. Therefore, the person skilled in the art would not have been led to use a catalyst comprising trivalent chromium in the claimed process.

- The Opposition Division made a wrong interpretation of the term "recycling" mentioned in document (1). "Recycling" was a broad term which was not limited to only describing the return of unreacted components to the original reaction vessel for further reaction but encompassed any system wherein one of the components is reused in some way, **unless** the context inevitably leads to a narrower interpretation. By way of examples the documents

(10) US-A- 4 579 998

(11) US-A- 4 066 423

(12) US-A- 4 983 273

were cited. Documents (10) and (12) disclosed processes wherein a component was recycled from a first reactor into a second reactor. In document (11), a portion of a product was recycled as absorbent.

- Furthermore, in the context of the disclosure of document (1), it was clear that the examples were not run continuously over a long period but rather the reaction was run discontinuously (i.e. the reaction was stopped after one run to allow evaluation of different catalysts). It was only in the context of discussing the discontinuously run examples that document (1) indicated at column 6, lines 16-21 that the by-product HCFC-133a produced was available for further reaction to produce the desired HFC-134a, by extending the catalyst contact time, raising the temperature, or recycling. The only other teaching regarding the further reaction of the HCFC-133a was at column 4, lines 22-27 where it was stated that the HCFC-133a "can be converted to the desired product either by further fluorination over the same catalyst system, or by using one of the catalysts known for this conversion". Column 6, lines 16-21 when read in the light of column 4, lines 22-27 made it clear that a broader meaning should be given to the term "recycling" in the context of document (1) than the definition given in document (16). Reading the whole of document (1) it was clear that, in the context of document (1), "recycling"

did not mean simply the return of the HCFC-133a to a single reaction zone but was a broad term intended to cover any re-use of the HCFC-133a to produce HFC-134a. Moreover, since HCl was produced during the conversion of HCFC-133a into HFC-134a, the person skilled in the art would have realized that reintroducing the product mixture into the reaction zone would have been detrimental and, therefore would have disregarded this option.

- Moreover, in view of the examples of document (1), the catalysts used did not appear particularly advantageous for the further fluorination of HCFC-133a to produce HFC-134a. Therefore, reading document (1), the person skilled in the art would have been led to conclude that it would be more suitable to treat unreacted HCFC-133a over a different catalyst system, known from the prior art as being suitable for the fluorination of HCFC-133a to form HFC-134a.

- The definition of the term "recycling" in document (16) was not the only meaning of this term that a person skilled in the art would consider when reading document (1). Documents

(17) Hackh's Chemical Dictionary, 4th Edition, McGraw-Hill, Inc., 1969, page 574;

(18) Grant and Hackh's Chemical Dictionary, 5th Edition, McGraw-Hill, Inc., 1987, page 499;

did not contain any entry for the terms "recycle" or "recycling". Presumably, the authors felt that these terms did not have a special meaning in the

field of chemistry. Documents

- (19) Webster's Ninth New Collegiate Dictionary, 1985, Merriam-Webster, Inc., page 985;
- (20) Kirk-Othmer Encyclopedia of Chemical Technology, 3rd Edition, 1982, John Wiley and Sons, Inc., pages 936-937

gave a definition of the term "recycle" as "reuse" or "recover". It was, therefore, clear from those documents that the definition of "recycling" given in document (16) was not the only definition of this term which would be known to a person skilled in the art in the chemical field.

- Even if it could be argued that the mention of recycle in document (1) would have been taken as referring to return of the HCFC-133a to the same reactor, there was no indication that trichloroethylene would be added at the same time as HCFC-133a was returned for further fluorination, given that the person skilled in the art could have expected that different reactional conditions would have been required depending on whether it was trichloroethylene or HCFC-133a that was being fed to the reactor.

VIII. The Respondent's submissions both in the written proceedings and at the oral proceedings can be summarised as follows:

- Document (1) disclosed a process for the reaction of trichloroethylene to HFC-134a involving a trivalent chromium as catalyst. It was clear for

the person skilled in the art that this process was continuous since feeding the gaseous mixture in a vertical nickel reactor tube and maintaining a residence time of 47 seconds described a continuous process. Furthermore, document (1) showed the possibility of recycling the intermediate HCFC-133a to the same reaction zone, obviously in presence of the feed trichloroethylene/HF since otherwise the process could not be continuous. Document (1) disclosed or suggested, therefore, all the features of the claimed process of each request.

- IX. The Appellant requested that the decision under appeal be set aside and that the patent be maintained on the basis as main request of Claims 1-12 as granted and a Claim 13 reading "The process of claim 1 wherein other organic by-products in said mixture are recycled", or as first auxiliary request of Claims 1-12 as granted, or as second auxiliary request on the basis of the claims submitted at the oral proceedings on 16 January 2002.

The Respondent requested that the appeal be dismissed.

- X. At the end of the oral proceedings the decision of the Board was announced orally.

Reasons for the Decision

1. The appeal is admissible.

Main request

2. *Article 123(2) and (3) EPC*

The sole amendment with respect to the set of claims as granted concerns the modification of Claim 13 (cf. point V above). This amendment is based on the application as filed, page 5, lines 33-34. The Board is, therefore, satisfied that Claim 13 does not contain subject matter which extends beyond the content of the application as filed. Nor does this Claim 13 which is dependent on Claim 1 in the form as granted extend the protection conferred. These points were not contested by the Respondent.

3. *Article 54(1)(2) EPC - Novelty*

3.1 A claimed invention lacks novelty unless it includes at least one technical feature which distinguishes it from the state of the art.

3.2 The Appellant argued that the subject matter of Claim 1 (cf. point II above) was new over the disclosure of document (1) since the latter did not disclose the same catalyst system.

According to Claim 1, the catalyst must meet two requirements:

- (a) it is "selected to form a mixture comprising 1,1,1,2-tetrafluoroethane and 2-chloro-1,1,1-trifluoroethane and less than 10 percent by weight of said other organic by-products"
- (b) it is "a catalyst composition comprising at least one of trivalent chromium, a Group VIII, Group VIIB, Group IIIB or Group IB metal and metals"

having an atomic number from 58 to 71".

However, those two features taken in combination do not by themselves ensure novelty of Claim 1 over the disclosure of document (1). Example No. 13 of document (1) describes a process for manufacturing 1,1,1,2-tetrafluoroethane (HFC-134a) by the reaction of HF and trichloroethylene (feeding of trichloroethylene at 0.293 g/min, column 4, lines 59-60; contact time 47 seconds, col.5, line 4) at 400°C in the presence of a **trivalent chromium** as catalyst ($\text{Cr}_2\text{O}(\text{OH})_4$ /coke) yielding HFC-134a, 2-chloro-1,1,1-trifluoroethane (HCFC-133a) and **less than 10%** by weight of organic by-products. This specific disclosure meets, therefore, the two conditions defining the catalyst system according to Claim 1.

3.3 By contrast, the recycling of HCFC-133a to the same reaction zone in a continuous process step where trichloroethylene and HF are continuously added is neither explicit nor necessarily implicitly disclosed in document (1). The subject matter of Claim 1 is, therefore, novel. For the same reasons, dependent claims 2 to 13 are also novel.

4. *Article 56 EPC - Inventive step*

4.1 The subject matter of Claim 1 relates to a process for the manufacture of 1,1,1,2-tetrafluoroethane (HFC-134a) by the reaction of HF and trichloroethylene, in the presence of a catalyst. This process is a continuous one by reason of the requirement that HCFC-133a be recycled to the reaction zone. Similar processes for the same objective belong to the state of the art: document (1) refers to a process for the manufacture of

HFC-134a which comprises contacting a haloethylene, preferably trichloroethylene, with HF, in a vapor phase reaction at elevated temperature, preferably 350°C to 550°C, in presence of a catalyst. This process is also continuous since trichloroethylene is fed at 0.293g/min, with a residence time of 30 to 300 seconds (cf. column 4, line 59 to column 5, line 4). The focus of said document lies on the use of a catalyst resulting from the codeposition of an hexavalent chromium oxide and a compound of a transitional metal selected from the group consisting of titanium, molybdenum and manganese, on alumina. However, it is not disputed that the disclosure of document (1) also describes as comparative tests two examples (Nos. 13 and 14) involving hydrated chromium oxide i.e. **trivalent chromium.**

The Board considers, in agreement with the parties, that document (1) represents the closest prior art, and hence takes it as the starting point in the assessment of inventive step.

- 4.2 The Appellant submitted that in view of this disclosure the technical problem to be solved might be seen in the provision of a process for economically manufacturing HFC-134a in a high amount and with less of organic by-products. However, in absence of evidence, the Board cannot acknowledge that the claimed process would provide any improvement compared to the process of document (1). In particular, the Board observes that some of the examples of this document produce a smaller amount of by-products (cf. Examples Nos. 4, 5 and 13) than the examples of the patent in suit. Thus, in view of these considerations, the technical problem as defined by the Appellant cannot be accepted by the

Board and consequently a reformulation of this technical problem becomes necessary to meet a less ambitious objective.

- 4.3 In the Board's judgment, having regard to the fact that there is no evidence for an improvement for all the claimed subject matter, the technical problem underlying the patent in suit in the light of the closest state of the art can only be seen in the provision of an alternative process for manufacturing 1,1,1,2-tetrafluoroethane.
- 4.4 In view of the technical information in the patent in suit, in particular in the examples, the Board is satisfied that the problem as defined in point 4.3 has been solved.
- 4.5 The remaining question is thus whether the prior art as a whole would have suggested to a person skilled in the art solving the technical problem indicated above in the way now claimed. In that context, the Board finds that the sole distinguishing feature of the claimed invention compared to document (1) is the **recycling** of the HCFC-133a to the same reaction zone in a continuous process where trichloroethylene and HF are continuously added (cf. point 3 above).
- 4.6 The Appellant argued in detail regarding the meaning of the term "recycling" in the context of document (1). The Board concurs that the meaning of this term is critical for the decision on inventive step in the present case. Document (1) discloses that "a particularly promising result shown in the Table is the high content of FC 133a in the impurities present in the product mixtures. This species is the last

intermediate before FC 134a and is available for further reaction either by extending the catalyst contact time, raising the temperature, or recycling" (cf. column 6, lines 16-21).

- 4.7 The Board considers that in the here relevant field of chemical process technology the term "recycling" is understood in accordance with the definition given in document (16), introduced by the Board as common general knowledge (cf. point VI above), i.e.

"the practice of returning a portion of the reaction products to the start of the system, either for the purpose of more efficient conversion of unreacted components or to reuse auxiliary materials that remain unchanged during processing".

The Appellant did not submit any documents which throw doubt on this being the meaning accepted in the field of chemical process technology. From the fact that documents (17) and (18), which are both Chemical Dictionaries, do not contain an entry for the terms "recycle" or "recycling" nothing can be deduced. The Board does not deny that there may exist another meaning for the term recycle, namely "reuse" (of wastes, for instance) as set out in documents (19) and (20) but these concern a different technical field far from the field here at issue. Documents (10), (11) and (12) are patents which do not normally form part of the common general knowledge, let alone the fact that document (12) is post published. Furthermore, contrary to the Appellant's view, in document (10), the recycling of paraffin through line 5 (cf. Figure 1) means returning it to the starting riser reactor (11) as set out on column 3, lines 9-16 and in document

(11), the term "recycling" is also used within the same meaning (cf. column 3, lines 53-55; column 4, lines 8-11, line 16, line 46 and line 56).

In conclusion, in the Board's judgment, the definition given in document (16) is the accepted meaning in the field of chemical process technology.

4.8 Nor is this conclusion affected by the presence of the passage of document (1), column 4, lines 22-29 stating that the major impurity in the product mixture is HCFC-133a which can be "converted to the desired product either by further fluorination over the same catalyst system, or by using one of the catalysts known for this conversion". This passage refers explicitly to documents (6) and (8) which state that unreacted organic starting materials i.e. HCFC-133a and by-products may be recycled to the process for further reaction to give the desired compound i.e. HFC-134a (cf. column 1, lines 61-65 and column 1, lines 60-64 respectively). The Board concludes that the expression "further fluorination over the same catalyst" includes the option of recycling to the starting reaction.

4.9 In that respect, the Board does not accept the view of the Appellant according to which the person skilled in the art would have been deterred from recycling the HCFC-133a due to the detrimental effect of HCl, since document (1) teaches that it is HCFC-133a which is available for recycling and not the whole mixture (cf. col.6, lines 16-21).

4.10 The Appellant also argued that, even though the person skilled in the art would have considered the option of recycling the HCFC-133a, he would have also noted that

this embodiment was envisaged in document (1) for processes involving hexavalent chromium catalyst and not trivalent chromium catalysts since the results given with the latter were given as comparative examples and the conversion and selectivity obtained were generally inferior to the examples within the definition of the process claimed in document (1). However, although this argument could have been taken into account if the technical problem had been to propose an improved process, it misses the point when only an alternative process is proposed. The person skilled in the art may derive from document (1) that, in view of Example No. 13 i.e. at 400 °C and in presence of a trivalent chromium ($\text{Cr}_2(\text{OH})_4/\text{coke}$), 6% of HFC-134a, 86% of HCFC-133a and 8% of other products are produced. This conversion cannot be regarded as significantly inferior to that of Example 5 using hexavalent chromium, for instance. Furthermore, seeking an alternative the person skilled in the art would have immediately observed that the process according to Example No. 13 produced as major component the HCFC-133a (86%). Therefore, in the Board's judgment, one of the possible alternatives to Example No. 13 offered to the person skilled in the art is to recycle the HCFC-133a, as taught by the same document in col.6, lines 16-21 (cf. point 4.6 above).

- 4.11 Faced with the technical problem of proposing an alternative process for the conversion of trichloroethylene to HFC-134a with a catalyst as disclosed in document (1), **in particular a trivalent chromium**, and as encompassed by the patent in suit, it would have been obvious, for the person skilled in the art to take into consideration the option of recycling the last intermediate HCFC-133a to the initial reaction

zone with trichloroethylene and HF. Furthermore, the additional features related to the presence of the feed trichloroethylene/HF in the ratio as defined in Claim 1 can only be regarded as an optimisation of the concentrations of each ingredients necessary to ensure the working of a continuous process. Indeed, introducing a molar amount of trichloroethylene at least equal to the molar amount of 1,1,1,2-tetrafluoroethane is a prerequisite condition and the stoichiometry of the reaction implies that at least four moles of HF for one mole of trichloroethylene be involved to get the HFC-134a. Complying with those features would thus be routine steps for the skilled person.

4.12 Therefore, in the Board's judgement, the subject-matter of Claim 1 of the main request represents a solution to the problem underlying the patent in suit which the skilled person would derive in an obvious manner from the prior art, and so does not involve an inventive step.

Since a decision can only be taken on a request as a whole, none of the further claims need to be examined.

4.13 Consequently, the main request has to be refused.

First auxiliary request

5. *Inventive step - Article 56 EPC*

Claim 1 of the first auxiliary request is the same as Claim 1 of the main request. For the same reasons as already set out in point 4 above, Claim 1 of the first auxiliary request does not involve an inventive step

and, therefore, this request must also fail.

Second auxiliary request

6. *Article 123(2) and (3) EPC*

Claim 1 results from the combination of the subject matter of Claims 1 and 3 as granted. The Board is satisfied that present Claim 1 does not contain subject matter which extends beyond the content of the application as filed. This Claim 1 is not amended so as to extend the protection conferred, either. This was not contested by the Respondent.

7. *Article 54(1)(2) EPC - Novelty*

The recycling of HCFC-133a to the same reaction zone in a continuous process step where trichloroethylene and HF are continuously added is neither explicitly nor necessarily implicitly disclosed in document (1). The subject matter of Claim 1 is, therefore, novel. For the same reasons, dependent claims 2 to 10 are also novel.

8. *Article 56 EPC - Inventive step*

8.1 Since present Claim 1 relates to a process involving a catalyst composition comprising a trivalent chromium, the Board finds that the reasons given for denying inventive step to Claim 1 of the main request apply *mutatis mutandi* to the present Claim 1 (cf. point 4 above). This request must also fail.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar:

The Chairman:

N. Maslin

P. P. Bracke